

# PATENT ABSTRACTS OF JAPAN

(11)Publication number : 10-237116  
 (43)Date of publication of application : 08.09.1998

(51)Int.Cl. C08F 2/48  
 C03C 27/06  
 C09K 3/10

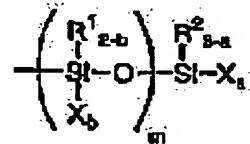
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## (54) CURABLE COMPOSITION

### (57)Abstract:

**PROBLEM TO BE SOLVED:** To obtain a curable composition, capable of providing a cured product having improved weather-resistant adhesion to various glasses and having high mechanical characteristics (hardness), by making the composition include a saturated hydrocarbon-based polymer having reactive silicon groups and a specify compound therein.

**SOLUTION:** This composition contains (A) a saturated hydrocarbon-based polymer, capable of providing a cured product having 2-6kg/cm<sup>2</sup> value of tensile stress at 50% elongation and having silicon-containing groups, having hydroxyl group or a hydrolyzable group bound to silicon group (e.g. an alkoxy group) and cross-linkable by forming a siloxane bond and represented by the formula [R<sup>1</sup> and R<sup>2</sup> are each a 1-20C alkyl, etc.; X is the hydroxyl group or the hydrolyzabl group; (a) is 0, 1, 2 or 3; (b) is 0, 1 or 2; (m) is 0 or 1-19:] (e.g. polyisobutylene) and (B) a photopolymerizable substance having 4 or more functional groups polymerizable by irradiation of light [e.g. (meth)acryloyl groups] in one molecule [e.g. pentaerythritol tetra(meth)acrylate]. The component B is preferably contained in an amount of 0.1-20pts.wt. based on 100pts.wt. component A having 7,000-15,000 molecular weight.



## LEGAL STATUS

[Date of request for examination]

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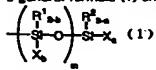
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## CLAIMS

## [Claim(s)]

[Claim 1] (A) a hardened material — 50 — % — tensile stress — a value — two — six — kgf/cm<sup>2</sup> — two — it is — silicon — an atom — having joined together — a hydroxyl group — or — hydrolysis — a sex — a machine — having — a siloxane — association — forming — things — constructing a bridge — obtaining — silicon — content — a radical — at least — one — a piece — having — saturated hydrocarbon — a system — a polymer — (— B —) — light — an exposure — a polymerization — carrying out — obtaining — a functional group — one — a molecule — inside — four — more than — having — photopolymerization — a sex — the matter — containing — things — the description — \*\* — carrying out — hardenability — a constituent.

[Claim 2] The silicon content radical which can construct a bridge by having the hydroxyl group or hydrolysis nature machine combined with the silicon atom, and forming siloxane association is a general formula (1) and [Formula 1].



(R1 and R2 are the Tori ORGANO siloxy radicals shown by the alkyl group of carbon numbers 1-20, the aryl group of carbon numbers 6-20, the aralkyl radical of carbon numbers 7-20, or (R')<sub>3</sub>SiO— (R' is the permutation or the unsubstituted hydrocarbon group of carbon numbers 1-20 independently, respectively) independently among a formula, respectively.) Moreover, X is a hydroxyl group or a hydrolysis nature machine independently, respectively. Furthermore, a is 0, 1, 2, or 3. b is 0, 1, or 2 and a and b are not simultaneously set to 0. moreover, m — the integer of 0, or 1-19 — it is — the hardenability constituent according to claim 1 which is the radical expressed.

[Claim 3] The hardenability constituent according to claim 2 characterized by X being an alkoxy group.

[Claim 4] The hardenability constituent according to claim 1 characterized by the number average molecular weight of a saturated hydrocarbon system polymer being 7000-15000.

[Claim 5] The hardenability constituent according to claim 1 characterized by the number average molecular weight per [ which can construct a bridge by having the hydroxyl group or hydrolysis nature machine combined with the silicon atom, and forming siloxane association ] silicon content radical being 4000-7000.

[Claim 6] The hardenability constituent according to claim 1 which is the polymer characterized by the total amount of the repeat unit to which a saturated hydrocarbon system polymer originates in an isobutylene having 50% of the weight or more.

[Claim 7] The hardenability constituent according to claim 1 with which the photopolymerization nature matter is characterized by being acrylate or methacrylate.

[Claim 8] (A) The hardenability constituent according to claim 1 characterized by containing 0.1-20 weight section for the (B) component to the component 100 weight section.

[Claim 9] The sealing material constituent for multiple glass characterized by containing the hardenability constituent of a publication in either of claims 1-8.

[Claim 10] The sealing material constituent for heat reflective glass characterized by containing the hardenability constituent of a publication in either of claims 1-8.

[Translation done.]

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## DETAILED DESCRIPTION

## [Detailed Description of the Invention]

[0001]

[Industrial Application] This invention relates to the hardenability constituent with which a useful weathering adhesive property and a useful hardened material degree of hardness have been improved by a sealing material or adhesives for the object for multiple glass, and a building etc.

[0002]

[Description of the Prior Art] It is known that the saturated-hydrocarbon system polymer which has the hydroxyl group or the hydrolysis nature machine combined with the silicon atom, and has at least one silicon content radical (henceforth "a reactant silicon radical") which can construct a bridge by forming siloxane association has the interesting property in which to construct a bridge by formation of siloxane association accompanied by the hydrolysis reaction of a reactant silicon radical etc., and a rubber-like hardened material is obtained with hygroscopic moisture etc. also in a room temperature. For this reason, it is effective if it uses for the sealing material for multiple glass, a structural elastic sealing compound, etc.

[0003] On the other hand, although especially the sealing material that uses for the circumference of glass, such as a sealing material for multiple glass, was asked for excellency in a weathering adhesive property, when the saturated hydrocarbon system polymer containing the reactant silicon radical of point \*\* was used, the weathering adhesive property was a little inadequate. Recently, many adhesives high heat reflective glass was used, and there was a problem that the weathering adhesive property over this heat reflective glass was especially inadequate.

[0004] Moreover, it was required to have the high modulus and high degree of hardness supporting the self-weight of glass in the sealing material for multiple glass unlike the structural sealing material which needs a low modulus and high elongation, and when the saturated hydrocarbon system polymer containing the reactant silicon radical of point \*\* was used, there was a problem that a mechanical characteristic (degree of hardness) was inadequate.

[0005]

[Problem(s) to be Solved by the Invention] This invention aims at raising the mechanical characteristic (degree of hardness) of a hardened material while it improves the weathering adhesive property over the various glass of the hardenability constituent which uses as a principal component the saturated hydrocarbon system polymer which has a reactant silicon radical.

[0006]

[Means for Solving the Problem] this invention person etc. improves the weathering adhesive property over various adherents, further, by finding out that hardened material physical properties (degree of hardness etc.) can be raised, solved said problem and completed this invention without doing adverse effects, such as lowering of the hardenability of this constituent, by adding a specific compound to this polymer, as a result of inquiring wholeheartedly, in order to solve such a problem.

[0007] Namely, the saturated hydrocarbon system polymer with which, as for this invention, the value of 50% tensile stress of the (A) hardened material has at least one reactant silicon radical

[0015] A reactant silicon radical may be in the end or the interior of a saturated hydrocarbon system polymer chain, and may be in both. Since the effective network chain density of the saturated hydrocarbon system polymer component contained in the hardened material formed eventually increases especially when a reactant silicon radical is in a molecule end, it is desirable from points, like the rubber-like hardened material of high elongation becomes easy to be obtained with high intensity.

[0016] moreover, the saturated hydrocarbon system polymer which has these reactivity silicon radical is independent — or two or more sorts can be used together. The polymer which makes the frame of a saturated hydrocarbon system polymer which has the reactant silicon radical used for this invention (1) [ whether it carries out a polymerization, using the olefinic compound of the carbon numbers 1-6, such as ethylene, a propylene, 1-butene, and an isobutylene, as the main monomer, and ] (2) Although it can obtain by the approach of hydrogenating after carrying out homopolymerization of the diene series, such as a butadiene and an isoprene, or carrying out copolymerization of the above-mentioned olefinic compound Since an isobutylene system polymer and a hydrogenation poly-butadiene system polymer tend to introduce a functional group into an end, and tend to control molecular weight and can make [ many ] the number of end functional groups, they are desirable.

[0017] the monomeric unit in which all the monomeric units may be formed in from the isobutylene unit, and an isobutylene system polymer has an isobutylene and copolymeric — an isobutylene system polymerization — the inside of the body is desirable and you may contain in 10% or less of range preferably especially 30% or less still more preferably 50% or less (below the same% of the weight). As such a monomer component, the olefin of carbon numbers 4-12, vinyl ether, an aromatic series vinyl compound, vinylsilane, and arylsilane are raised, for example. As such a copolymer component, for example 1-butene, 2-butene, A 2-MECHIRU 1-butene, a 3-MECHIRU 1-butene, a pentene, a 4-MECHIRU 1-pentene. A hexene, a vinyl cyclohexene, the methyl vinyl ether, ethyl vinyl ether, Isobutyl vinyl ether, styrene, alpha-methyl styrene, dimethyl styrene, Monochlorostyrene, dichloro styrene, beta-pinenene, an indene, vinyl trichlorosilane. Vinyl methyl dichlorosilane, vinyl dimethyl chlorosilane, vinylidinemethylmethoxysilane. A vinyl trimethyl silane, divinyl dichlorosilane, divinyl dimethoxysilane, Divinyl dimethylsilane, 1, 3-divinyls 1, 1, and 3, 3-tetramethyl disiloxane, TORISI methylsilane, a tetra vinyl silane, allyl trichlorosilane, Allyl compound methyl dichlorosilane, allyl compound dimethyl chlorosilane, allyl compound dimethyl methoxysilane, An allyl compound trimethyl silane, diaryl dichlorosilane, diaryl dimethoxysilane, diaryl dimethylsilane, gamma-methacryloyl oxypropyl trimethoxysilane, gamma-methacryloyl oxypropyl dimethoxysilane, etc. are raised.

[0018] Moreover, as an isobutylene and a monomer which has copolymeric, if vinylsilane and arylsilane are used, the radical which a silicon content increases and can act as a silane coupling agent will increase, and the adhesive property of the constituent obtained will improve. Other monomeric units may be made to contain besides the monomeric unit used as a principal component like the case of the above-mentioned isobutylene system polymer in a hydrogenation poly-butadiene system polymer or other saturated hydrocarbon system polymers.

[0019] moreover, a monomeric unit by which double association after a polymerization like polyene compounds, such as a butadiene and an isoprene, remains in the saturated hydrocarbon system polymer which has the reactant silicon radical used for this invention in the range in which the object of this invention is attained — small quantity — you may make it contain in 1% or less of range 5 more% or less 10% or less especially preferably Preferably, as for the number average molecular weight of an isobutylene system polymer or a hydrogenation poly-butadiene system polymer, it is desirable a saturated hydrocarbon system polymer and that it is 7,000 to about 15,000, and its about 8,000 to 12,000 Equifused thing which is, carried out, and has a fluidity is especially desirable from points, such as handling and a cone.

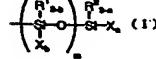
[0020] Moreover, an isobutylene system polymer or a hydrogenation poly-butadiene system polymer has a saturated hydrocarbon system polymer and the preferably desirable thing which has the ratio (Mn/Fn) of the number (Fn) of the number average molecular weight per reactant silicon radical, i.e., number average molecular weight, (Mn), and the reactant silicon radicals per molecule within the limits of 4000-7000. It is because the value of 50% tensile stress of the

which is 2-6 kgf/cm<sup>2</sup>. (B) It is a thing about the hardenability constituent characterized by containing the photopolymerization nature matter which has the functional group which can carry out a polymerization by the exposure of light four or more in 1 molecule. As opposed to the saturated hydrocarbon system polymer 100 weight section which contains at least one reactant silicon radical especially in the molecule whose value of 50% tensile stress of the (A) hardened material is 2-6 kgf/cm<sup>2</sup> and whose molecular weight is 7000-15,000 (B) It is related with the hardenability constituent with which the weathering adhesive property characterized by containing the acrylate or the methacrylate 0.1-20 weight sections which have the functional group which can carry out a polymerization by the exposure of light four or more in 1 molecule has been improved.

[0008]

[Embodiment of the Invention] Hereafter, this invention is explained in detail. The saturated hydrocarbon system polymer which has the reactant silicon radical used for this invention is a polymer which does not contain carbon-carbon unsaturated bonds other than a ring substantially, for example, polyethylene, polypropylene, a polyisobutylene, hydrogenation polybutadiene, hydrogenation polyisoprene, etc. are raised. As a reactant silicon radical, it is a general formula (1) and [0009].

[Formula 2]



[0010] (R1) and R2 are the Tori ORGANO siloxy radicals shown by the alkyl group of carbon numbers 1-20, the aryl group of carbon numbers 6-20, the aralkyl radical of carbon numbers 7-20, or (R) 3SiO — (R' is the permutation or the unsubstituted hydrocarbon group of carbon numbers 1-20 independently, respectively) independently among a formula, respectively. Moreover, X is a hydroxyl group or a hydrolysis nature machine independently, respectively. Furthermore, a is 0, 1, 2, or 3, b is 0, 1, or 2 and a and b are not simultaneously set to 0, moreover, m — the integer of 0, or 1-19 — it is — the radical expressed is raised.

[0011] As a hydrolysis nature machine, the radical currently generally [ a hydrogen atom, an alkoxy group, an acyloxy radical, a KETOKISHI mate radical, the amino group, an amide group, an amino oxy-radical, a sulfhydryl group, an alkenyloxy radical, etc. ] used, for example is raised. Among these, although an alkoxy group, an amide group, and an amino oxy-radical are desirable, the point of hydrolysis nature being mild and being easy to deal with it to especially an alkoxy group is desirable.

[0012] A hydrolysis nature machine and a hydroxyl group can be combined with one silicon atom in the 1-3 range, and a (s+sigma) has 1-5 desirable range. When a hydrolysis nature machine and a hydroxyl group join together in [ two or more ] a reactant silicon radical, they may be the same and may differ. Although the silicon atom which forms a reactant silicon radical is one or more pieces, in the case of the silicon atom connected by siloxane association etc., it is desirable that they are 20 or less pieces. Especially, it is a general formula (2).

[0013]

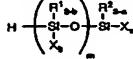


[0014] Since the reactant silicon radical expressed with (the inside of a formula, and R2, X and a are the same as the above) is easy to receive, it is desirable. The reactant silicon radical in a saturated hydrocarbon system polymer 1 molecule is one or more pieces, and it is desirable that there are 1,1-5 pieces. If the number of the reactant silicon radicals contained in a molecule becomes less than one piece, hardenability becomes inadequate and good rubber elasticity may not no longer be obtained.

hardenability of a base polymer is determined by the number average molecular weight per reactant silicon radical, i.e., the molecular weight between the points constructing a bridge. When the number average molecular weight per reactant silicon radical is larger than 7000, the value of tensile stress may not become two or more 2 kgf/cm 50% and when the number average molecular weight per reactant silicon radical is less than 4000, the value of tensile stress may not become two or less 6 kgf/cm 50%.

[0021] The process of the saturated hydrocarbon system polymer which has a reactant silicon radical next is explained. the end organic-functions mold obtained by the polymerization method (cationic polymerization method using the specific compound combining and [ which is called inphor ] and a chain transfer agent) the isobutylene system polymer which has a reactant silicon radical at the chain end among the isobutylene system polymers which have a reactant silicon radical is called inphor method — it can manufacture preferably using all end organic-functions mold isobutylene system polymers. For example, the general formula after obtaining the polyisobutylene which has a partial saturation radical at the end by the partial saturation radical installation reaction to a polymer which is indicated by the dehydrohalogenation reaction and JP,63-105005,A of this polymer etc. [0022]

[Formula 4]



[0023] (— R1, R2, X, a, and b are the same as the above among a formula) — the hydroxilane compound (this compound is a compound which the hydrogen atom combined with the radical expressed with a general formula (1)) expressed — desirable — general formula [0024]

[Formula 5]



[0025] (— R2, X, and a are the same as the above among a formula) — the approach of introducing a reactant silicon radical into a polymer is raised by carrying out the addition reaction called a hydroxilane reaction using a platinum catalyst in the hydroxilane compound expressed. As a hydroxilane compound, for example Trichlorosilane, methyl dichlorosilane, The halogenation silanes like dimethyl chlorosilane and phenyl dichlorosilane; Trimethoxysilane, Triethoxysilane, methyl diethoxysilane, methyl dimethoxysilane, The alkoxysilane like phenyl dimethoxysilane; A methyl diaxotropy silane, The acyloxy silane like a phenyl diaxotropy silane; although the KETOKISHI mate silanes like bis(dimethyl KETOKISHI mate) methylsilane and bis(cyclohexyl KETOKISHI mate) methylsilane are raised, it is not limited to these. Especially among these, halogenation silanes and alkoxysilane are desirable.

[0026] Such a manufacturing method is indicated by each description of JP,4-69859,B, JP,7-108928,B, JP,63-254149,A, JP,64-22904,A, and the patent official report No. 2539445 etc. Moreover, the isobutylene system polymer which has a reactant silicon radical inside a chain is manufactured by making the vinylsilane and arylsilane which have a reactant silicon radical add and copolymerize in the monomer which makes an isobutylene a subject.

[0027] Furthermore, after making the vinylsilane and arylsilane which have a reactant silicon radical in addition to the isobutylene monomer which is a principal component copolymerize on the occasion of the polymerization at the time of manufacturing the isobutylene system polymer which has a reactant silicon radical at the chain end, the isobutylene system polymer which has a reactant silicon radical inside an end and a chain is manufactured by introducing a reactant silicon radical into an end.

[0028] As the vinylsilane which has a reactant silicon radical, and arylsilane For example, vinyl trichlorosilane, vinyl methyl dichlorosilane, Vinyl dimethyl chlorosilane, Vinylidemethylmethoxysilane, divinyl dichlorosilane, Divinyl dimethoxysilane, allyltrichlorosilane, allyl

compound methyl dichlorosilane, Allyl compound dimethyl chlorosilane, allyl compound dimethyl methoxysilane, diaryl dichlorosilane, diaryl dimethoxysilane, gamma-methacryloyl oxypropyl trimethoxysilane, gamma-methacryloyl oxypropyl methyl dimethoxysilane, etc. are raised. [0029] Said hydrogenation poly-butadiene system polymer is the hydroxyl group of an end hydroxy hydrogenation poly-butadiene system polymer first, for example. - It is general formula after making it oxy-metal radicals, such as ONa and -O.K.,  $(2\text{CH}_2=\text{CH}-\text{R}_2\text{Y})$ . (3) (For the inside of a formula, and Y, halogen atoms such as a chlorine atom and an iodine atom, and R2 are  $\text{R}_2\text{Y}$ ,  $-\text{R}_2\text{OCO}$ , or  $-\text{R}_2\text{CO}$  ( $\text{R}_2$  is the divalent hydrocarbon group of carbon numbers 1-20)) an alkylene group, a cyclo alkylene group, an alkene radical, and an alkylene group — being desirable — by the divalent organic radical shown  $-\text{CH}_2-$  and  $-\text{R}'-$  the divalent radical chosen from  $-\text{CH}_2\text{H}-\text{CH}_2-$  ( $\text{R}'$  is the hydrocarbon group of carbon numbers 1-10) — especially — being desirable — the hydrogenation poly-butadiene system polymer which has an end olefin radical is manufactured by making the organic halogenated compound shown react.

[0030] The approach to which it is made to react with alkali hydroxides such as Na, metal alkoxide, NaOH like metal hydride NaOCH3 like alkali-metallocal H like K, and KOH, etc. as an approach of making the end hydroxyl group of an end hydroxy hydrogenation poly-butadiene system polymer an oxy-metal radical is raised. Although an end olefin hydrogenation poly-butadiene system polymer with the almost same molecular weight as the end hydroxy hydrogenation poly-butadiene system polymer used as a start raw material is obtained by said approach. Before making the organic halogenated compound of a general formula (3) react to obtain the polymer of the amount of macromolecules more, a methylene chloride, bis(chloromethyl) benzene, the bis(chloromethyl) ether, etc. solve. If it is made to react with the organic halogenated compound which molecular weight can be increased if it is made to react with the multiple-valued organic halogenated compound which contains two or more halogens in 1 molecule, and is shown by the general formula (3) after that it can deal in the hydrogenation poly-butadiene system polymer which is the amount of giant molecules more, and has an olefin radical at the end.

[0031] As an example of the organic halogenated compound shown by said general formula (3), although an allyl chloride, an allyl compound star's picture, vinyl (chloromethyl) benzene, allyl compound (chloromethyl) benzene, allyl compound (bromomethyl) benzene, the allyl compound (chloromethyl) ether, allyl compound (chloromethoxy) benzene, 1-butene (chloromethyl) ether, 1-hexene (chloromethoxy) benzene, allyoxy (chloromethyl) benzene, etc. are raised, for example, it is not limited to them. Among these, an allyl chloride is cheap, and since it moreover reacts easily, it is desirable.

[0032] Installation of the reactant silicon radical to said end olefin hydrogenation poly-butadiene system polymer is manufactured by carrying out an addition reaction using a platinum system catalyst in a hydrosilane compound like the case of the isobutylene system polymer which has a reactant silicon radical at the chain end. As for the saturated hydrocarbon system polymer which has at least one reactant silicon radical which is the (A) component of this invention, it is important that the value of 50% tensile stress of a hardened material which stiffened the polymer is  $2 - 6 \text{ kgf/cm}^2$ . Here, the 50% tensile stress of a hardened material is as follows.

[0033] namely, the saturated hydrocarbon system polymer 100 section (the same the weight section and the following) which has at least one reactant silicon radical — receiving — the water 1 section, the cyclic acid tin 3 section, and a lauryl amine — 0.75 \*\*\*\*\*. It slushes into shattering with a thickness of 3mm, after often mixing and carrying out degassing. It is recuperated for four days at 50 more degrees C for three days by 23 degrees C in this thing, and the hardened material sheet of about 3mm thickness is obtained. From this sheet, the No. 3 dumbbell based on JIS-K6301 is pierced, and a tensile test is performed (tension rate 500 mm/min). The tensile strength to 50% ductility in this case is defined as tensile stress 50%.

[0034] In this invention, the value of the above-mentioned 50% tensile stress uses only the thing of  $2 - 6 \text{ kgf/cm}^2$  as a saturated hydrocarbon system polymer (A) among the saturated hydrocarbon system polymers which have at least one reactant silicon radical. When a degree of hardness may become inadequate when the values of 50% tensile stress are less than two 2

constituent, and the weathering adhesive property which come to contain the saturated hydrocarbon system polymer with which it has a reactant silicon radical when the number of said functional groups uses four or more photopolymerization nature matter for the constituent of this invention are impractical. Although the loadings of the photopolymerization nature matter have the 0.1 - 20 desirable section to the (A) component 100 section, its 1 - 10 section is still more desirable, and especially its thing to do for 2-5 section combination is desirable. When loadings are the less than 0.1 sections, the improvement effect of hardened material physical properties such as a degree of hardness, and a weathering adhesive property is not sometimes enough and the 20 sections are surpassed, the storage stability of this hardenability constituent may fall.

[0041] Various additives are added by the hardenability constituent of this invention if needed. As an example of such an additive, the physical-properties regulator which adjusts the tractive characteristics of the curing catalyst which promotes a silanol condensation reaction, and the hardened material to generate, a plasticizer, a bulking agent, an adhesive improver, an antioxidant, radical inhibitor, an ultraviolet ray absorbent, a metal deactivator, anti-ozonant, light stabilizer, the Lynn system peroxide decomposition agent, lubricant, a pigment, a foaming agent, etc. are raised, for example.

[0042] The example of such an additive is indicated by each description of JP-4-69659B, JP-7-108928B, JP-63-254149A, and JP-64-22904A etc. When it uses for various glass, such as general-purpose inorganic glass (float glass), a remarkable weathering adhesive property improvement effect is shown, but the hardenability constituent of this invention has especially the remarkable effectiveness of improving a weathering adhesive property, when it is used as a sealing material constituent for heat reflective glass.

[0043] The above-mentioned heat reflective glass shows glass equipped with the optical function of reflecting or absorbing the light of specific wavelength by covering a metal membrane, the metal nitride film, the metallic-oxide film, etc. on a glass front face. The effectiveness of the photopolymerization nature matter of this invention is similarly accepted, when the various above-mentioned additives are added. That is, when the hardenability constituent of this invention is used for sealing materials for direct grading, such as a sealing material for multiple glass, a sealing material for speed signal generator, methods of construction, and an automobile, these sealing materials' mechanical characteristic and weathering adhesive properties (degree of hardness etc.) can be improved by addition of this photopolymerization nature matter.

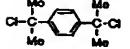
[0044] [Example] Although an example and the example of a comparison explain this invention concretely below, this invention is not limited to this.

[0045]

[The example 1 of manufacture] After acting the nitrogen purge of the inside of installation and a container to a 500ml proof-pressure glass container as a three-way cock, ethylcyclohexane (what was dried by leaving 1 or more night with molecular-sieves 3A) 54ml and toluene (what was dried by leaving 1 or more night with molecular-sieves 3A) 126ml, and p-DCC(following compound) 0.76g (3.28mmol) were added to it in the container using the syringe.

[0046]

[Formula 6]



[0047] Next, proof-pressure glass liquefied gas extraction tubing with a needle valve with which close was connected to the three-way cock, and after isobutylene monomer 56ml attached the polymerization container all over -70 degrees C dry ice / ethanol bus and cooled, it made the inside of a container reduced pressure using the vacuum pump. After opening the needle valve and introducing an isobutylene monomer in a polymerization container from liquefied gas extraction tubing, the inside of a container was returned to ordinary pressure by introducing

kg/cm, and surpassing  $6 \text{ kgf/cm}^2$ , \*\* of the maximum elongation may become a small weak hardened material.

[0035] When the saturated hydrocarbon system polymer which has a reactant silicon radical as mentioned above does not contain substantially the unsaturated bond which is not a ring in a molecule, compared with the sealing agent which consists of a conventional rubber system polymer like an organic system polymer or an oxy-ethylene system polymer which has an unsaturated bond, weatherability becomes good remarkably. Moreover, since this polymer is a hydrocarbon system polymer, moisture cutoff nature and its water resisting property are good, and it becomes the low hardened material of moisture cutoff nature while it has the adhesive ability which was excellent to various mineral base materials, such as glass and aluminum. Furthermore, in this invention, in order that the value of tensile stress may use the polymer of  $2 - 6 \text{ kgf/cm}^2$  50%, it becomes the hardened material which are a high modulus and a high degree of hardness, and has flexible rubber elasticity.

[0036] 10% or more of the content of the saturated hydrocarbon system polymer which has a reactant silicon radical in the hardenability constituent of this invention is desirable, is more desirable, and is desirable, [ especially 30% or more of ] [ 20% or more of ] in the hardenability constituent of this invention, while raising a hardened material degree of hardness further, in order to raise a weathering adhesive property, the photopolymerization nature matter is used as a (B) component. By irradiating light, this photopolymerization nature matter is a compound which has the partial saturation radical which causes a polymerization reaction, when the double bond of intramolecular is activated. Various things such as a constituent containing an organic monomer, oligomer, resin, or these, are known by this kind of matter, and the matter of commercial arbitration can be used in the invention. (A) When the photopolymerization nature matter is added as a (B) component for a component, it is thought that the (B) component acts as a weathering adhesive property amelioration agent of the (A) component by the exposure of light since a hard coat can be formed in respect of adhesion with adherends, such as glass. However, early bond strength does not fall.

[0037] In the monofunctional case in which only the one number of the sensitization radicals (partial saturation radical) of the above and the photopolymerization nature matter is, a linear polymer is formed according to photopolymerization. However, since the polymer molecule of the network structure is formed since photopolymerization and optical bridge formation take place simultaneously in the case of the polyfunctional photopolymerization nature matter which has two or more sensitization radicals (partial saturation radical), and a hard coat can be formed according to an adhesion interface, a weathering adhesive property and a mechanical characteristic (degree of hardness) are improved. The improvement effect of a weathering adhesive property and a mechanical characteristic is so large that it has many sensitization radicals (partial saturation radical) of the photopolymerization nature matter, in order to improve these properties notably, the number of functional groups has four or more desirable pieces, and its five or more pieces are still more desirable.

[0038] As an example of representation of the photopolymerization nature partial saturation radical contained in this photopolymerization nature matter, although a vinyl group, an allyl group, a vinyl ether radical, a vinyl thioether radical, the vinyl amino group, an acetylene nature partial saturation radical, an acryloyl radical, a methacryloyl radical, a styryl radical, a cinnamoyl radical, etc. can be mentioned, the acrylate which makes an acryloyl radical or a methacryloyl radical a sensitization radical also in these (meta) is more desirable from the reason of optical initiation effectiveness being easy to come to hand highly. In addition, in this description, acrylate (meta) names acrylate and methacrylate generically.

[0039] The number of functional groups is the monomer which has acrylic or four or more methacrylic system partial saturation radicals, oligomer, or such mixture as four or more acrylate (meta), and can mention pentaerythritol tetraprop (meta) acrylate, dipentaerythritol PENTA, hexa (meta) acrylate, etc. as an example of a monomer. Moreover, as an example of oligomer, with a molecular weight [ such as polyethylene GURIKORUJI (meta) acrylate and polypropylene GURIKORUJI (meta) acrylate, ] of 10000 or less oligo ester can be mentioned.

[0040] Hardened material physical properties, such as a degree of hardness of a hardenability

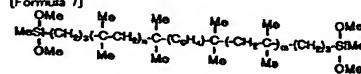
nitrogen from one side in a three-way cock. Next, 2-methylpyridine 0.112g (1.2mmol) was added. Next, in addition, the polymerization was started 1.65ml (15.1mmol) of titanium tetrachlorides. After [ of reaction time ] 70 minutes, allyl compound trimethyl silane 0.89g (7.9mmol) was added, and the introductory reaction of an allyl group was performed at the polymer end. After 200ml of water washed the reaction solution 4 times after [ of reaction time ] 120 minutes, the allyl compound end isobutylene system polymer was obtained by distilling off a solvent.

[0048] Subsequently, allyl compound end isobutylene polymer obtained in this way — After dissolving in n-heptane 20ml and carrying out temperature up of the 40° to about 70 degrees C, methyl dimethoxysilane 1.5g (0.01mmol) and the platinum (vinyl siloxane) complex  $1 \times 10^{-4}$  (eq/vinyl group) were added, and the hydrolysis reaction was performed. FT-IR performed the reaction trace and olefin absorption of  $1640\text{cm}^{-1}$  disappeared in about 4 hours.

[0049] By carrying out vacuum concentration of the reaction solution, isobutylene polymer — which has a reactant silicon radical in the target both ends was obtained. (The following compound)

[0050]

[Formula 7]



[0051] In this way, while computing yield from the yield of the obtained polymer, Mn and Mw/Mn were calculated by measuring the reinforcement of the resonance signal of a proton (the proton of the initiator origin: 6.5-7.5 ppm, methyl proton: 0.0-0.1ppm combined with the silicon atom of the polymer end origin, and methoxy proton: 3.4-3.5) which belongs to each structure by 300MHz 1H-NMR analysis, and comparing end structure by the GPC method, again. 1H-NMR is Varian. It measured in CDCl3 using Gemini300 (300MHz for 1H).

[0052] In addition, FT-IR — the Shimadzu make — IR-408 and GPC — as a liquid-sending system — Waters LC Module1 and a column — Shodex It carried out using K-804. Molecular weight is given with the relative molecular weight to a polystyrene standard. The analysis values of a polymer were  $Mn=17500$ ,  $Mw/Mn=1.14$ , and  $Fn(\text{sil})=1.91$ . (For number average molecular weight, polystyrene conversion and the number of end silyl functional groups are the number per isobutylene polymer 1 molecule).

[0053]

[The example 2 of manufacture] The isobutylene system polymer which has a reactant silicon radical like the example 1 of manufacture was compounded except having changed the addition into p-DCC1.16g (0.20mmol), 2-methylpyridine 0.093g (1.0mmol), and allyl compound trimethyl silane 1.22g (10.8mmol).

[0054] The analysis values of a polymer were  $Mn=11400$ ,  $Mw/Mn=1.23$ , and  $Fn(\text{sil})=1.76$ .

[0055]

[An example 3 and the example 1 of a comparison] From each analysis value of the base polymer compounded in the examples 1 and 2 of manufacture, the ratio ( $Mn/Fn$ ) of the number average molecular weight per reactant silicon radical, i.e., number average molecular weight, ( $Mn$ ), and the reactant silicon radicals per molecule was calculated. The value of  $Mn/Fn$  is shown in a table 1.

[0056] Next, 50% tensile stress of the hardened material of the base polymer compounded in the examples 1 and 2 of manufacture was measured. A result is shown in a table 1. In addition, the measuring method of 50% tensile stress is based on the following approach as it was described previously, namely, the base polymer 100 section — receiving — the water 1 section, the cyclic acid tin 3 section, and a lauryl amine — 0.75 \*\*\*\*\*. It slushes into shattering with a thickness of 3mm, after often mixing and carrying out degassing. It is recuperated for four days at 50 more degrees C for three days by 23 degrees C in this thing, and the hardened material sheet of about 3mm thickness is obtained. From this sheet, the No. 3 dumbbell based on JIS-K6301 was pierced, and the tensile test was performed (tension rate 500 mm/min). Tensile strength to 50%

